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Chemical Constituents of Some Endemic Conifers in Taiwan

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Chemical constituents of conifers of families of Cupressaceae, Pinaceae and Taxodiaceae, of which endemic species such as *Chamaecyparis taiwanensis, Chamaecyparis formosensis, Calocedrus formosana, Pinus armandii, Tsuga chinensis* and *Taiwania cryptomerioides* are emphasized. The chemical constituents mainly include terpenes, lignans and flavones. Methods and significance of this research are discussed. Extraction, crystallization and chromatography are general methods for separation of plant constituents. Chemical and spectral methods including IR, MS and NMR are frequently used for structural determination along with X-ray crystallography and circular dichroic spectroscopy to establish absolute configuration in appropriate cases. Finding hinokitiol from Taiwan cypress has initiated the emerging study of troponoid chemistry and nonbenzenoid aromatic compounds. A norsesquiterpene chamaecynone and a mixture of sesquiterpenes T-cadinol and T-muurolol exhibit respectively termiticide and antifidant activities. The flavonoid and stilbene constituents are analyzed to support the chemotaxonomy of armand pine and Taiwan white pine. A study of the constituents of armand pine indicates that 30-*nor*-3β-methoxyserrat-14-en-21-one is biogenetically derived from 3β-methoxyserrat-14-en-21-one by an oxidative degradation via intermediacy of the 30-alcohol and the 30-aldehyde.

INTRODUCTION

Located in the subtropical area and with the greater part of the island formed by a massive mountain chain attaining a height over 2500 m, Taiwan has abundant alpine plants. The main components of the coniferous forests include several valuable species, such as Taiwan cypress (台灣扁柏), Taiwan red cypress (紅檜), Taiwan incense cedar (肖楠), Ranta fir (巒大杉), and Taiwania (台灣杉). These have been important sources of commercial timber and pharmacologically active substances. Taiwan forest resource postage stamps were recently released to publicize the beauty of forests in Taiwan and bring to public attention the conservation of these woods.

Research of natural products has been active in Taiwan. The scope of natural product research related to chemistry, pharmacology and other subjects in Taiwan has been described by Y. H. Kuo.[1,2] Here we review chemical constituents of conifers of families of Cupressaceae, Pinaceae and Taxodiaceae, of which endemic species are emphasized.

CUPRESSACEAE

Taiwan red cypress, *Chamaecyparis formosensis* Matsum., is a giant endemic species with small scale-like leaves and elliptic cones, which grows in the mountain areas at elevations 900-2600 m. The primary leaves grow on the lower stem and branches of seedlings. Taiwan cypress, *Chamaecyparis taiwanensis* Masamune & Suzuki, is a close relative of Taiwan red cypress with similar characteristics except round cones. Although the Taiwan red cypress (benihi in Japanese), Taiwan cypress (taiwanhinoki in Japanese) and Japanese hinoki (*Chamaecyparis obtusa* Sieb. et Zucc.) look similar, the chemical constituents of these plants differ. T. Nozoe began the study of components of essential oil of Taiwan cypress when he established the research laboratory at Taihoku Imperial University (in Taipei), which was renamed National Taiwan University after repatriation of Taiwan to China in 1945. He isolated an enolic compound, hinokitiol, from the heartwood of Taiwan cypress and showed its molecular formula $C_{10}H_{12}O_2$ in 1938. As the spectral instruments, which are extensively used currently, were then unavailable, the structural elucidation relied on chemical methods which generally required a large sample and work over a protracted period. The research was interrupted by World War II. Because cypress oil can be used as a substitute for motor fuel, the Japanese army ordered the Taipei factory of Takasago Perfumery Company to manufacture the oil from old stumps of cypress trees. At the end of the war, T. Nozoe helped to reorganize the laboratory in the chemistry department but encountered difficulty in obtaining re-

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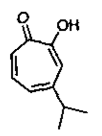
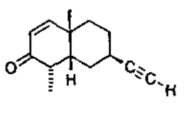
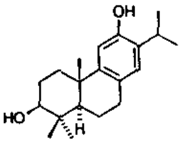
This review is invited to commemorate the sixtieth anniversary of the Chinese Chemical Society.

search materials. Fortunately, he found the 'red mud' containing Taiwan cypress oil that was accidently dumped in the backyard of the Takasago Perfumery Company during the wartime. Because Taiwan cypress oil contains 0.1-0.2% of hinokitiol, the red mud yielded several kilograms of hinokitiol crystals that one had never imagined. The structure of hinokitiol was finally determined to be 2-hydroxy-4-isopropyl-2,4,6-cycloheptatrienone (1) in 1948.[3] Owing to the intrinsic property of the polarizing carbonyl group and the resulting intramolecular hydrogen bonding, this seven-membered cyclotrienolone structure is best delineated as a form of tropylium ion which was an aromatic compound of new type. The emerging study of troponoid chemistry and nonbenzenoid aromatic compounds was thus initiated.[4]


1, hinokitiol
(β-thujaplicin)


2, (-)-chamaecynone


3, (+)-hinokiol

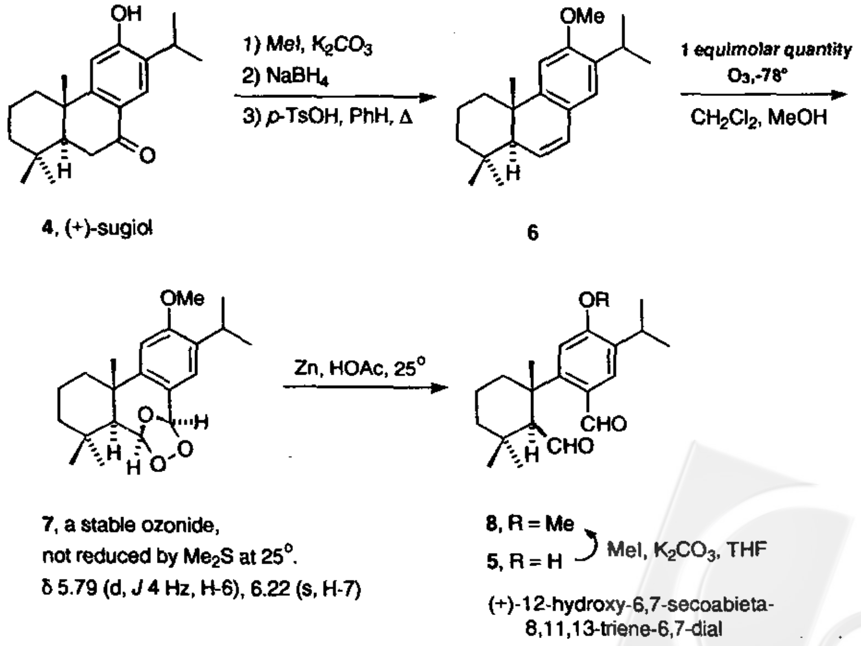
The study of chemical constituents in plants of *Chamaecyparis* genus was continued by Y. T. Lin and coworkers.[5-9] In a search for an improved method to separate phenolic components, the technique of polyamide thin-layer chromatography was introduced by K. T. Wang after 1960.[10-13] For this finding one was greatly indebted to Y. T. Lin for he had sacrificed a precious imported nylon shirt that was cut into pieces and soaked in *N,N*-dimethylformamide for the first preparation of polyamide thin layers.

Taiwan red cypress is known to possess strong resistance against wood-decaying fungi.[21-25] From the essential oil, Y. S. Cheng isolated a norsesquiterpene ($C_{14}H_{18}O$) chamaecynone (2), which is the first natural product found to contain an acetylenic group[14] unlike the isopropyl group in common sesquiterpenes. This compound shows the termiticide activity but causes no significant toxic effect on human beings or domestic animals.[21,22] Six congeners of chamaecynone were isolated later.[15-20]

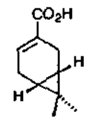
In addition to abietane-type diterpenoids of hinokiol (3)[26] and sugiol (4),[27] an uncommon secoabietane dialdehyde (5) was isolated from the bark.[26] Since 1960, spectral methods were gradually adopted in determination of molecular structures. However, many natural products chemists still considered bench experiments to be the ultimate approach. We carried out a chemical correlation between the dialdehyde (5) and sugiol (Scheme I). A stable diterpenoidal ozonide (7) was prepared during this course;[28] this ozonide is inert to Me_2S at room temperature and stable on standing in $CDCl_3$ for over 50 days.

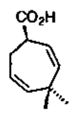
Taiwan incense cedar, *Calocedrus formosana* Florin (previously named *Libocedrus formosana* Florin), is commonly called 'shonan' in Taiwan. It is an evergreen species with scale-like leaves and long-ovoid cones which grows in mountain areas at elevations 300-2000 m. The chemical constituents of the wood, bark and leaves have been exten-

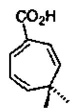
Scheme I

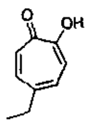


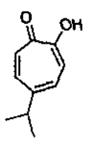
sively investigated.[29-45] The wood contains a large quantity of terpenoidal acids. A bicyclo[4.3.0]heptene carboxylic acid, chaminic acid (**9**), was first reported in 1960. The wood also contains many seven-membered cyclic compounds, such as shonanic acid (**10**), thujic acid (**11**), 5-ethyltropolone (**12**), *β*-thujaplicin (**1**), *γ*-thujaplicin (**13**) and *β*-thujaplicinol (**14**). Monoterpenes and five cadinane-type sesquiterpenes have been identified.[42] Diterpenoidal components in the wood predominate in abietane-type phenols,[40] such as hinokiol (**3**), sugiol (**4**), 1-oxo-hinokiol (**15**), shonanol (**16**), ferruginol (**17**), 6*α*-hydroxy-7-oxo-ferruginol (**18**), and xanthoperol (**19**), whereas labdane-type acid derivatives,[43] such as communic acid (**20**), isocupressic acid (**21**), and pinosolide acid (**22**) are rich in the leaves. A 1,3-dioxototarol (**23**), an uncommon norlabdenoic acid (**24**)[48] and a labdadienol conjugated with coumaric acid (**25**) have also been isolated. Lignans, including hinokinin (**26**), savinin (hibalactone, **27**), 7-oxohinokinin (**28**), calocedrin (**29**), matairesinol (**30**), 4′,5-dihydroxy-3,3′,4-trimethoxylign-7-en-9,9′-olide (**31**) and 4,4′-dihydroxy-3,3′,5-trimethoxylign-7-en-9,9′-olide (**32**), were obtained and their structures were chemically correlated. For instance, the lignan dihydroanhydride, (+)-calocedrin was reduced with $NaBH_4$ in an alkaline condition to give a racemic mixture of savinin.[41] Compound **28** exists as the keto but not enol form.[45] Lignans are compounds of a class derived from two propylbenzene units jointed by the 8,8′-carbons. The naturally occurring epoxylignans generally have 7,7′- and 7,9′-linkages, *e.g.* in **26**-**32**, but **33** (named as shonanin) is a rare 9,9′-epoxylignan.[44] The known lignans with 9,9′-linkage typically occur as higher oxidative derivatives of lactones and hemiacetals.

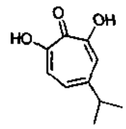

9, (+)-chaminic acid

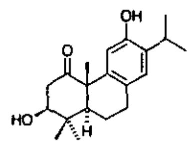

10, (-)-shonanic acid

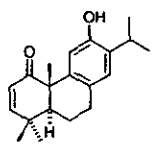

11, thujic acid

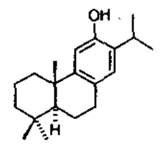

12, 5-ethyltropolone

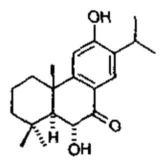

13, γ-thujaplicin

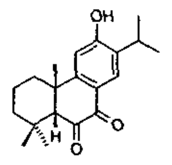

14, β-thujaplicinol

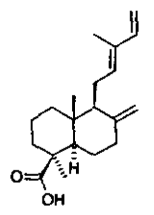

15, (+)-1-oxohinokiol

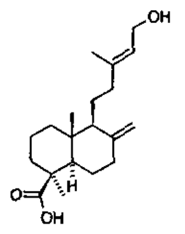

16, (+)-shonanol

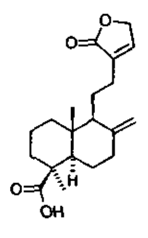

17, ferruginol

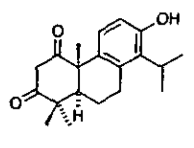

18, 6α-hydroxy-7-oxo-ferruginol

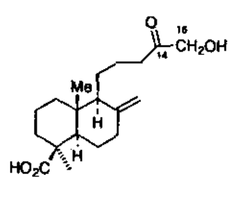

19, xanthoperol

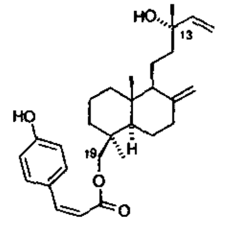

20, *trans*-communic acid

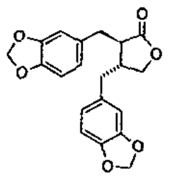

21, isocuppressic acid

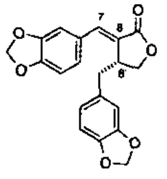

22, pinosolide acid

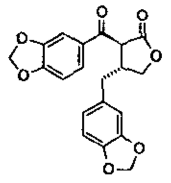

23, (+)-1,3-dioxototarol

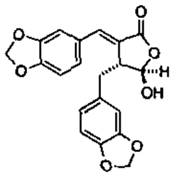

24, 15-nor-16-hydroxy-14-oxolabd-8(17)-enoic acid

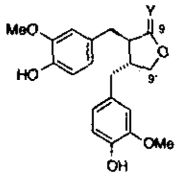

25, 13*S*-labda-8(17),14-diene-19-(*cis*-*p*-hydroxycinnamoxy)-13-ol

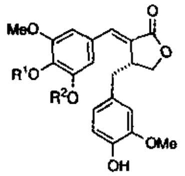

26, (-)-hinokinin


27, (-)-savinin or hibalactone


28, 7-oxohinokinin


29, (+)-calocedrin

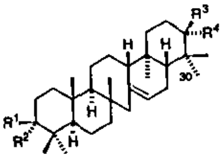

Y=
30, (-)-matairesinol O
33, 4,4′-dihydroxy-3,3′-dimethoxy-9,9′-epoxylignan H, H


R¹= R²=
31 Me H
32 H Me

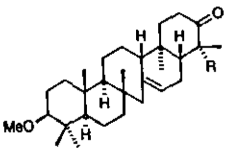
PINACEAE

After 1970, we isolated several serratene crystals (**34**-**38**) from *Pinus luchuensis* Mayer[46,47] (琉球松), which is an introduced species but popularly cultivated in Taiwan. Serratene is a kind of pentacyclic triterpene bearing a central seven-membered C ring, a double bond between C-14 and C-15, and seven tertiary methyl groups instead of eight methyls in common pentacyclic triterpenes. An ^1H NMR spectrum would clearly reveal these characteristic structural features. However, the National Taiwan University was equipped only with a 60-MHz NMR instrument, which generally required a sample of large concentration. Because deuterated solvents were too expensive and not readily available, we had to use hot CCl_4 to prepare the sample solution. When the operator kindly informed us of readiness to record the spectra, one author (J. M. Fang) carried the sample solution, by dipping the tube in a beaker containing hot water to prevent precipitation of crystals, and rush to the machine for a quick measurement. We recently found a series of serratenes from an endemic conifer,[51] *Pinus armandii* Franchet var. *masteriana* Hayata (台灣華山松). Thanks to Taiwan's rapid economic growth, our department, like other institutes, benefits from advanced spectral instruments, mostly provided by the National Science Council and the Ministry of Education. A student is currently able to record a satisfactory spectrum on a superconducting FT-NMR instrument even with a minute sample in $CDCl_3$ solution. Finding the serratenone (**34**), the alcohol (**39**), the aldehyde (**40**) and the norserratenone (**41**) in the same plant source, *P. armandii*, indicates that 30-norserratenone (**41**) is biogenetically derived from the serratenone (**34**) by a process of oxidative degradation via the intermediacy of (**39**) and (**40**).

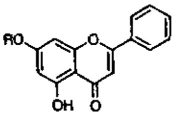
The armand pine, *P. armandii*, is morphologically considered a variety of Taiwan endemic *P. morrisonicola* Hayata (台灣五葉松) and Japanese white pine, *P. parviflora* Sieb. et Zuuc. The flavonoid and stilbene components were analyzed to support the chemotaxonomy of the three species (Table 1).[48-51] Chrysin (**42**) is the most abundant flavone whereas *trans*-pinosylvin monomethyl ether (**48**) is the major stilbene. Other major components include tectochrysin (**43**), pinocembrin (**44**), pinostrobin (**45**), pinobanksin (**46**) and *trans*-pinosylvin dimethyl ether (**49**). Two reports concern the terpenoid and serratenoid constituents of *Pinus taiwanensis* (台灣二葉松).[54,55]



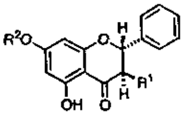
	R¹=	R²=	R³=	R⁴=
34	OMe	H	=O	
35	OH	H	=O	
36	=O		=O	
37	OMe	H	H	OMe
38	OMe	H	H	OH



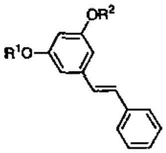
	R=
34	CH_3
39	CH_2OH
40	CHO
41	H



	R=
42, chrysin	H
43, tectochrysin	Me



	R¹=	R²=
44, pinocembrin	H	H
45, pinostrobin	H	Me
46, pinobanksin	OH	H



	R¹=	R²=
47, *trans*-pinosylvin	H	H
48, monomethyl ether	Me	H
49, dimethyl ether	Me	Me

The Taiwan hemlock, *Tsuga chinensis* Pritzel var. *formosana* Hayata (台灣鐵杉), is a major forest tree indigenous to the high mountain area (2000-3500 m). Many related lignans were isolated and their structures were determined by chemical and spectral methods.[56,58] Undoubtedly X-ray crystallography is the most efficient method for structural determination, especially when many chiral centers are involved.[59-62] However, there are many

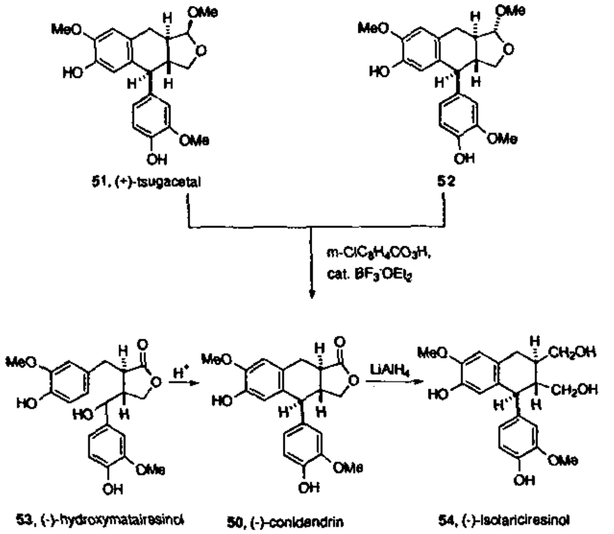
Table 1. Flavonoids and stilbenes in the heartwood of *P. armandii, P. morrisonicola* and *P. parviflora*

Compound	*P. armandii*	*P. morrisonicola*	*P. parviflora*
chrysin	* (9.51)	* (50.10)	*
tectochrysin	* (0.59)	* (3.75)	-
pinocembrin	* (2.61)	* (0.15)	*
pinostrobin	* (0.18)	* (0.26)	-
pinobanksin	* (2.85)	* (3.13)	*
pinobanksin 3-acetate	* (3.56)	-	-
strobopinin	-	-	*
cryptostrobin	-	-	*
trans-pinosylvin	-	* (0.09)	*
trans-pinosylvin monomethyl ether	* (42.80)	* (38.87)	*
trans-pinosylvin dimethyl ether	* (14.20)	* (0.22)	-
dihydropinosylvin	* (0.15)	-	*
dihydropinosylvin monomethyl ether	* (19.02)	-	*
dihydropinosylvin dimethyl ether	* (0.35)	* (0.08)	-

*Indicates presence and - indicates absence of the specified compound. The number in parentheses indicates relative percentage content.

instances when the compounds are inherently noncrystalline. In such cases, there is no alternative but to utilize various spectral techniques and chemical methods. In Scheme II, we show the lignans are chemically correlated with α-conidendrin (**50**), a commonly found lignan lactone. The stereochemistry of tsugacetal (**51**) is established by X-ray analysis, whereas its C-9 epimer (**52**) had previously isolated from *Dacrydium intermedium* Kirk, Podocarpaceae. The chemical contituents of the wood of *Keteleeria davidiana* Beissner var. *formosana* Hayata (台灣油杉), Pinaceae, were reported.[63]

Scheme II



Although X-ray diffraction is a powerful method to determine the relative configurations of molecules, it is not pertinent to solve the absolute configurations of those organic molecules containing no heavy atom. As subjection to derivatization with a known chiral reagent often risks the uncrystallizable products useless for X-ray analysis, utilization of circular dichroism (or optical rotatory dispersion) appears to be a reasonable alternative method, especially in the cases abiding by the exciton coupling theory (or the Cotton effect). CD spectroscopy was successfully applied to determine the absolute configurations of the flavones from *P. morrisonicola*[53] and the lignans from *Juniperus chinensis* Linn. var. *kaizuka* Hort (龍柏),[64] Cupressaceae, a common ornamental tree in Taiwan.

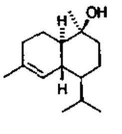
TAXODIACEAE

Taiwania, *Taiwania cryptomerioides* Hayata (台灣杉), is an endemic evergreen species with thick linear-triangular leaves and elongate ovoid cones. The chemical constituents of this plant have been investigated exhaustively.[65-83] Several cadinane-type sesquiterpenes were isolated from the essential oil of the wood.[69-77] By using preparative gas chromatography Y. S. Cheng separated T-cadinol (**55**) and T-muurolol (**56**) from a mixture which had previously been considered a homogeneous substance denoted χ-cadinol found in the essential oil of Taiwan red cypress.[19,69] T-cadinol is a sesquiterpene alcohol with the bicyclo-[4.4.0]decene skeleton, whereas T-muurolol is the isomer having a different chirality at the C-10 center. Their structures are distinguished by means of ^{1}H NMR spectra, in which the vinyl proton of T-cadinol occurs as a singlet whereas that of T-muurolol appears as a doublet (J = 5 Hz). A mixture of these two compounds is found to exhibit a deterrent effect against the Taiwan red-bellied tree squirrel.[86] The components of lignans and biflavones were also reported.[78,80-82]

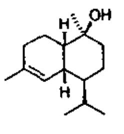
Ranta fir, *Cunninghamia konishii* Hayata, is characterized by flat leaves, elliptic cones and linear-falcate foliage with finely toothed margins; its chemical constituents have not been thoroughly investigated.[84,85] A study of the essential oil of the bark indicated the presence of caryophyllene (**57**) and caryophyllene oxide (**58**); however, the content of (**58**) increased at the expense of (**57**) as shown by the GC analysis. This phenomenon indicates that caryophyllene oxide is an artifact possibly produced by autoxidation of caryophyllene. Related projects on the autoxidation and microbial oxidation of some terpenes were subsequently carried out.[87-92]

REMARKS

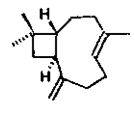
Research on natural products in Taiwan has a span of nearly 60 years, almost coinciding with the establishment of the Chinese Chemical Society. At least one hundred papers have been published on chemical constituents of endemic conifers in Taiwan. We cannot definitely evaluate the impact of this work on the academic society, but it can be considered a representative endeavour to which many people have contributed even in a harsh research environment. With the development of many sophisticated spectral instruments, structural elucidation has become quite 'trivial'; however, the isolation and purification of natural compounds are never 'routine' work. In retrospect, we agree with K. Nakanishi.[93] Natural-product chemistry traditionally meant a branch of organic chemistry in which one isolated compounds from plants, animals, microorganisms etc., and carried out structural studies and, in many cases, syntheses. In the classical sense, structure determination and synthesis were generally the termination of a natural product project; but this condition is no longer the case. Structural determination of a bioactive compound is simply the starting point for the next step, namely clarification of the structural interaction between a small substrate molecule and its receptor biopolymer. Natural product chemists, because of their training in isolation, purification, and structural determination, are absolutely indispensable. As research facilities are greatly improved, chemists in Taiwan should accept the challenge to treat 'dynamic' natural products that exist in only minute quantities, or which may have only a fleeting existence. This task requires unselfish devotion and interdisciplinary collaboration of scientists to attain this objective.



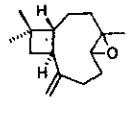
55, (-)-T-cadinol



56, (-)-T-muurolol



57, β-caryophyllene



58, β-caryophyllene oxide

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Key Words

Cupressaceae; Pinaceae; Taxodiaceae; *Chamaecyparis taiwanensis*; *Chamaecyparis formosensis*; *Calocedrus formosana*; *Pinus armandii*; *Pinus luchuensis*; *Pinus morrisonicola*; *Tsuga chinensis*; *Taiwania cryptomerioides*; *Cunninghamia konishii*; Terpenes; Lignans; Flavones.

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